

Correlation between conductivity and structural evolution of Si-rich silicon oxide films annealed at different temperatures

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Si/SiO₂ quantum dot (QD) nanostructures are considered as viable basic building blocks in future all-Si tandem solar cells. Moreover, they are of potential use for new heterocontacts and passivation layers of silicon-based wafer and thin-film solar cells.

Essential prerequisites for the photovoltaic application of such QDs are precise control of their structural, interfacial and electrical properties. Here we present an X-ray photoelectron spectroscopy (XPS) analysis of the formation of ultrathin layers of Si/SiO₂ QDs under ultrahigh vacuum conditions by self-organized growth from thermally deposited silicon-rich silicon oxide (SiO_x, x<2) layers, which are thermodynamically unstable and therefore undergo phase separation upon appropriate *in situ* post-annealing. Moreover we establish a correlation of the structural evolution of the QDs and the corresponding electrical properties revealed by atomic force microscopy (AFM) techniques.

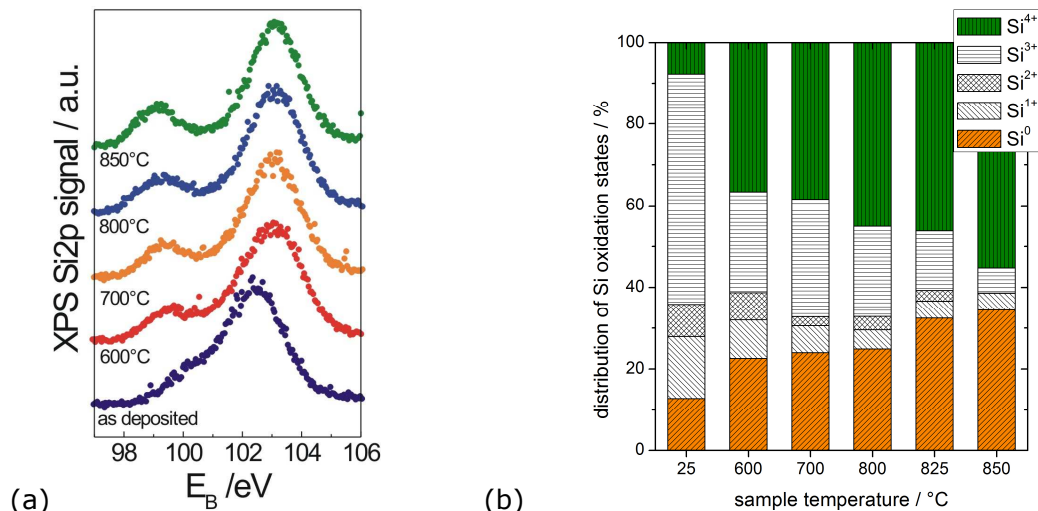


Fig. 1: (a) XPS Si 2p spectra of a 10 nm SiO_x layer in the initial state (25°C) and after subsequent vacuum annealing up to 850°C. (b) Amounts of Si oxidation states in the layer as deposited and after annealing.

Fig. 1a shows the spectra of the Si 2p core levels for a 10 nm SiO_x layer at different stages of vacuum annealing up to 850°C. Phase separation started below 600°C and was completed at around 850°C. By controlling layer thickness and stoichiometry, QD density and size were adjusted, allowing for a tuning of the quantization energies and of the interlayer transport properties [1]. Quantitative information about the contributions of the individual oxidation states to the Si 2p signal is obtained by curve decomposition according to the procedure given in Ref. [2]. Fig. 1b shows the relative amounts of the constituting Si suboxides (i.e., Si in different oxidation states). Both, the relative concentrations of elemental Si (i.e., Si⁰) and of silicon dioxide (i.e., Si⁴⁺) increase monotonically by subsequent increase of the annealing temperature. The relative concentrations of the Si suboxides decrease

monotonically and reaches a total concentration ($\text{Si}^{1+} + \text{Si}^{2+} + \text{Si}^{3+}$) of less than 10% of the total signal at 850°C. Thus, phase separation is completed and Si QDs embedded in SiO_2 are formed with only residual amounts of suboxides remaining at the Si QD / SiO_2 interfaces

The AFM topograph of an ultrathin Si/ SiO_2 QD layer (Fig. 2a) reveals slight protrusions and indicates the formation of QDs. The Kelvin force microscopy (KFM) image (Fig. 2b) clearly visualizes dot-like structures and shows true material contrast because the potential is not directly correlated to topography and the potential differences are rather high (~ 1 V) [3].

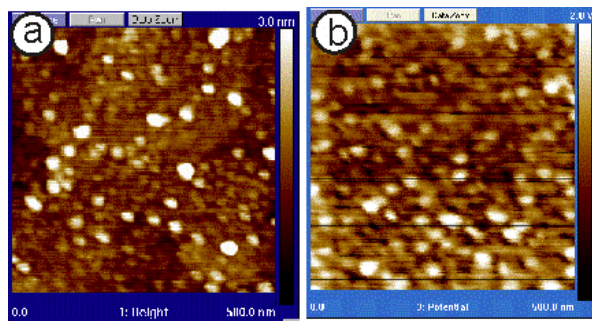


Figure 2: (a) tapping mode AFM topograph of the 10 nm Si/ SiO_2 QD layer, z-range: 3 nm. (b) KFM image (different position), z-range: 2.0 V. Scanning size: 500 × 500 nm².

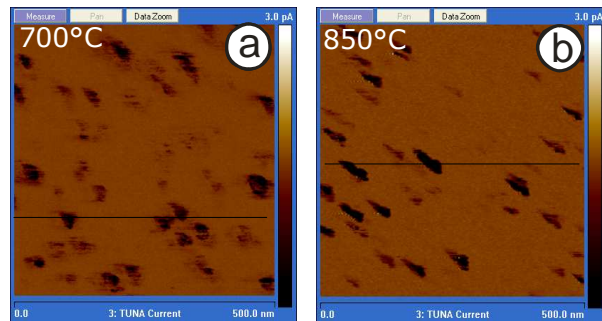


Figure 3: Current-sensing AFM images of the 10 nm SiO_x layer upon (a) 700°C and (b) 850°C annealing. Scanning size: 500 × 500 nm².

Spatially resolved information on the electronic structure of the QD layer is obtained by applying a bias voltage between the AFM tip and the sample and sensing the resulting current (CS-AFM). Typical CS-AFM images for the samples annealed at 700°C and 850°C are shown in Fig. 2a and 2b, respectively. The positions of the darker (i.e., more conductive) areas correlate with the dots in the simultaneously recorded topography maps. A clear correlation of increasing QD conduction with increasing annealing temperature and proceeding phase separation is observed. Increasing the annealing temperature from 700 °C up to 850 °C leads to an increase of the current by a factor of >5 due to the completed expelling of oxygen atoms and expansion of the QDs. The discrepancy in the number of QDs is just due to the number of smaller QDs being invisible in the KFM images. In contrast, on the as-deposited SiO_x precursor layer no current (i.e., <0.05 pA) was detected even under maximum bias applied (± 10 V).

In conclusion, the gradual phase separation revealed by XPS is directly correlated with electrical properties as derived from AFM measurements detecting local conductivities and surface potentials across individual QDs. CS-AFM images confirm QD formation and prove their electrical conductivity which reaches its maximum when phase separation is completed.

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